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16. ABSTRACT

Measurements from the Regional Air Pollution Study in St. Louis and from synoptic rawinsonde stations in North America were used with annual point and area source emission data in St. Louis to establish some consequences of local and large-scale ozone transport.

In rural areas outside of St. Louis, ozone concentrations exceeded the National Ambient Air Quality Standard of 80 ppb (160 $\mu g/m^3$) and could not be attributed to the emissions of pollutants within the metropolitan area of St. Louis. Typically, these high ozone concentrations occurred when the air flowing into St. Louis had been associated with an anticyclone during the 3 days prior to its arrival. Trajectories indicated that during these 3-day periods the air had remained within the eastern half of the United States where there are numerous high-intensity urban-industrial centers. On other days during the study analyses of the local air flow confirmed that the urban-industrial areas within the immediate vicinity of St. Louis were responsible for high ozone concentration in the city as well as in the rural areas. An analysis of the formation and movement of an "ozone cloud" through the network of stations revealed the consequences of local ozone generation and transport.

17. KEY WORDS AND DOCUMENT ANALYSIS

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OZONE TRANSPORT IN THE ST. LOUIS AREA

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Abstract—A network of 25 stations in and around St. Louis, Missouri, continuously recorded wind speed, wind direction, and ozone concentration near the surface as part of the Environment Protection Agency's Regional Air Pollution Study. Data were also obtained on an hourly basis from several pilot-balloon sites within the St. Louis area. These measurements, from July 1 to September 15, 1975 and from May 1 to June 19, 1976, were used with annual point and area source emission data in St. Louis to establish some consequences of the local transport of ozone generated within St. Louis. In addition, these data were analyzed in conjunction with synoptic rawinsonde measurements from North America to establish the significance of large-scale ozone transport into the St. Louis area.

In rural areas outside of the city ozone concentrations exceeded the National Ambient Air Quality Standard of 80 ppb (160 $\mu g\ m^{-3}$) and could not be attributed to the emissions of pollutants within the metropolitan area of St. Louis. During the period of study this occurred on 15 days when meteorological conditions were conducive to the large-scale transport of ozone. Typically, these high ozone concentrations occurred when the air flowing into St. Louis had been associated with an anticyclone during the 3 days prior to its arrival. Trajectories indicated that during these 3-day periods the air had remained within the eastern half of the United States where there are numerous high-intensity urban-industrial centers. On other days during the study, analyses of the local air flow confirmed that the urban-industrial areas within the immediate vicinity of St. Louis were responsible for high ozone concentration in the city as well as in the rural areas. An analysis of the formation and movement of an "ozone cloud" through the network of stations revealed the consequences of local ozone generation and transport.

1. INTRODUCTION

Ozone transport† occurs across hundreds of kilometers and crosses regional and national boundaries. This phenomenon has recently received considerable attention (Cleveland and Kleiner, 1975; Lyons and Cole, 1976; Cox *et al.*, 1975). The frequency of large-scale ($\geq$ 200 km) transport of ozone into metropolitan areas and its contribution to the ozone burden of an area has become an issue of major concern. Insufficient knowledge about ozone transport masks the source(s) responsible for locally measured concentrations, and thereby hinders the effectiveness of control strategies for protection against undesirable levels of this contaminant.

A comprehensive set of measurements, made by the U.S. Environmental Protection Agency's (EPA) Regional Air Pollution Study (RAPS) in St. Louis, Missouri, were analyzed in an attempt to understand the phenomenon in that region. The data were collected from a network of 25 continuously recording Regional Air Monitoring System (RAMS) stations. Additional measurements were used from four pilot-balloon (PIBAL) stations in the RAPS and from the North American network of rawinsonde stations. The data selected for analysis were recorded from July 1 to September 15, 1975 and from May 1 to June 19, 1976 – periods during which high ozone concentrations were common in St. Louis.

2. DATA AND STATION SITES

Wind speed, wind direction, and ozone concentrations in St. Louis were measured continuously near ground level. To obtain wind speed and direction aloft, PIBALs were routinely taken each hour (weather permitting) at two stations during weekdays. During the time period from July 6 to August 15, 1975 hourly PIBALs were taken every day of the week at four stations.

Ozone was measured using the chemiluminescence technique with the intakes at 4 m above ground level. Cup anemometers and wind vanes used in the RAMS were very sensitive with low starting thresholds (Myers and Reagan, 1975) and were mounted on top of 10 or 30 m towers.

Figure 1 depicts the locations of the 25 RAMS stations and the four PIBAL stations. The rural or outer sites, 121–125, were all located in undeveloped or agricultural areas. A criterion for the placement of each station was that there could be no significant obstruction to the airflow higher than one-tenth the distance from the obstruction to the measurement point (Poller, 1974). All the stations were situated such that the land use was homogeneous for at least 2 km around each site.

* On assignment from the National Oceanic and Atmospheric Administration, U.S. Department of Commerce.

† "Ozone transport" and "the transport of ozone and its precursors" are used interchangeably.

Fig. 1. Locations of pilot-balloon stations (141–144) and the Regional Air Monitoring System stations (101–125). Areas of significantly high NO_x and total hydrocarbon emissions are enclosed by broken lines.

One-minute averages of the data, which were automatically recorded on magnetic tape at RAPS headquarters, were checked by various computer programs for erroneous values (Jurgens and Rhodes, 1976). Tests used to screen the data consisted of continuity and relational checks as well as the operational mode of the instruments. Even so, visual inspection revealed some highly suspicious and unreasonable values which were deleted from further study.

3. ANALYSIS PROCEDURES

3.1 *Emissions within the St. Louis area*

Each area associated with significantly high emissions of total hydrocarbons (THC) and nitrous oxides (NO_x) was outlined (Fig. 1) by broken lines and was termed a "high emission area" (HEA). The HEAs were derived from point and area annual emission rates in EPA's National Emissions Data System (1973 National Emissions Report, 1976). The criteria for HEAs were: (1) in total they include at least two-thirds of all the THC and NO_x annual emissions within a 100 by 100 km area centered on St. Louis and (2) they include only those areas with high annual emission rates of NO_x and THC per unit area (emission rates per unit area which were greater than the emission rate per unit area of the 100 by 100 km area). Although the HEAs in Fig. 1 enclose only one-eighth of the 100 by 100 km area around St. Louis they account for approximately three-fourths of all the THC emissions and two-thirds of all the NO_x emissions. Considering average emission rates per unit area, the HEAs emit NO_x 13 times faster and THC 20 times faster than the remainder of the 100 by 100 km area.

3.2 *Locally generated ozone transported within St. Louis*

Local-scale trajectories were calculated backward in time from each outer station, 121–125, for which the hourly average ozone concentration exceeded the 1 h National Ambient Air Quality Standard (NAAQS) of 80 ppb (160 $\mu g\,m^{-3}$). If, on a given day, the standard was exceeded for more than 1 h, the time of the highest ozone concentration was used as the starting time in the trajectory calculations. These trajectories are referred to as "local-scale trajectories" since they are limited to the local wind data recorded at the 10 and 30 m towers in the RAMS. Each local-scale trajectory was computed backward in time until one of the following conditions was met: (1) the trajectory intersected one of the HEAs, (2) the trajectory was more than 100 km from the nearest RAMS site, or (3) 24 h had elapsed since the trajectory began. Statistics were compiled relating the number of times the local-scale trajectories intersected the HEAs to the ozone concentrations observed at the outer RAMS sites. On a particular day in August during 1975 relatively high ozone concentrations occurred at several RAMS stations. This situation was investigated in greater detail than usual in regard to the generation and transport of ozone within the RAMS network. Using data from PIBAL sites and the local-scale trajectory calculations originating at different times from several RAMS stations, the development and movement of an "ozone cloud" was detected.

3.3 *Ozone transported into St. Louis*

Of special interest in this portion of the study were the days when the ozone measured at the outer sites

could not be attributed to the emissions within the HEAs. The local-scale trajectories were used for this purpose. PIBAL analyses were also utilized to further substantiate the local-scale trajectories. From the PIBAL analyses statistics were derived regarding the direction, speed, and range of the wind within the lowest 1 to 2 km of the planetary boundary layer. These statistics were compiled for approximately 36 h prior to the time when the NAAQS for ozone was exceeded at stations 121–125. Ozone data from stations in and close to the HEAs were not included in any aspect of this portion of the study due to the difficulty involved in separating locally generated ozone and ozone transported into the area from distant sources.

Three-day backward-in-time large-scale trajectories were computed using 6 hourly* North American rawinsonde data. These trajectories originated at one of the rural sites, 121–125. The trajectories were of special interest on those days when the following criteria were met: (1) the NAAQS was exceeded at any of the rural sites, 121–125, (2) the local and large-scale trajectories did not interesect a HEA, (3) analysis of PIBAL data revealed that it was unlikely that the wind would transport air from a HEA to the rurual station of interest, and (4) points back in time along a local-scale trajectory continually increased their distance from a HEA. The method of calculating the large-scale trajectories was developed by Heffter and Taylor (1975). Winds within the 50 to 1500 m layer above the terrain were used in the trajectory calculations.

4. COMPUTATIONAL TECHNIQUES

4.1 *Local-scale trajectories*

Each trajectory consisted of a series of 10 min segments calculated from hourly average vector wind speeds and directions. For example, the 10 min segment back from 1059 to 1050 was based on the hourly average winds during 1000 to 1059. Likewise, the trajectory segment from 1009 back to 1000 was based on the same hourly average winds. Segments consisted of a weighted average wind based on measurements at nearby RAMS stations. Each wind reported within the scanning radius, R (Fig. 2), was weighted by an r^{-2} weighting factor where r is the distance between the mid-point of a trajectory segment and a station within the scanning radius. Iteration was necessary to determine the distance of a station within the scanning radius to the mid-point of the trajectory segment. The scanning radius was equal to 10 km at the center of St. Louis, but for every 2 km away from the center of the city it increased by 1 km. If there were fewer than three stations within the scanning radius, it was increased by increments of 2 km until three stations were within its radius. The initial choice of a 10 km scanning radius in the city center was made because at this distance only urban as opposed to rural stations would influence the trajectory segment. It was necessary to incorporate a variable scanning radius because of the disparity in the density of the RAMS sites.

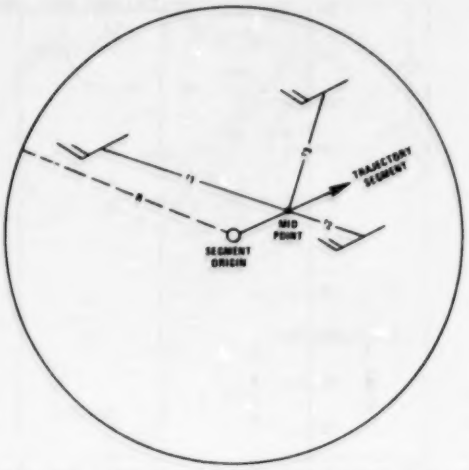

Fig. 2. Illustration of the utilization of the parameters in the local-scale trajectory calculations.

4.2 *Large-scale trajectories*

For a complete discussion of the large-scale trajectory technique the reader is referred to Heffter and Taylor (1975). The major computational difference between the large- and local-scale trajectories is the inclusion of a term to account for the properties of the low-level jet in the large-scale trajectories. To calculate large-scale trajectories at least two reporting stations must be within 556 km (300 nm) of a segment origin or one station must be within 278 km (150 nm) of a segment origin. A 6 to 9 h persistence in the winds is considered acceptable for input into a trajectory segment. Beyond 9 h persistence is not assumed, and if there are too few stations within the segment origin the trajectory calculations terminate. The time interval is 3 h, after which new stations (because of a new trajectory segment endpoint) may influence the length and direction of the next trajectory segment.

4.3 *Wind statistics based on PIBAL data*

The range of a data sample often incorporates outliers in the data and with respect to the wind incorporates variability in wind direction without regard to the wind speed. For these reasons a weighted range statistic was defined for the PIBAL measurements. Two expressions are needed to define the weighted range (WR) of the wind:

$$\frac{\sum_{i=1}^{m} V_i}{\sum_{i=1}^{n} V_i} \geq \tau \qquad (1)$$

and

$$WR = 2\theta_{i=m} \qquad (2)$$

* The vast majority of observations however, were at 12 h intervals.

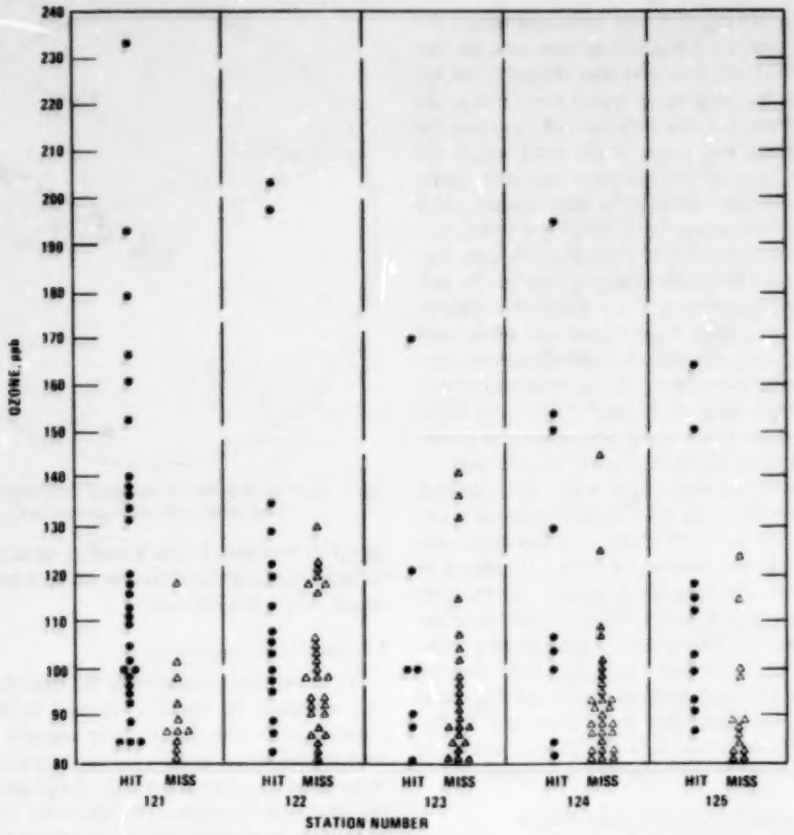

Fig. 3. Ozone concentration categorized with respect to the local-scale trajectory either intersecting (hit) or missing a "high emission area".

Table 1. Contingency table relating ozone concentrations at outer RAMS sites to whether or not the local-scale trajectory intersected a "high emission area"

[O_3] \ Emission	Hit	Miss	Total
80–89 ppb	14	44	58
90–105 ppb	19	36	55
> 105 ppb	40	19	59
TOTAL	73	99	172

where θ_i is the deviation of an observed wind direction from the resultant vector wind direction, V_i is an observed scalar wind speed, τ is predefined such that $0 < \tau \leq 1$, n is the number of observations in the sample, and m is equal to the first number less than or equal to n which satisfies expression (1). Each θ_i and its associated V_i is sorted such that $\theta_i \leq \theta_{i+1}$. When $\tau = 1$ the weighted range is identically equal to the range. A value of 0.95 was chosen for τ because of its similarity to twice the s.d. Ninety five percent of the area under a normal curve is contained within twice the standard deviation.*

*The S.D. is applicable to continuous functions only, and therefore could not be applied to θ_i.

5. RESULTS AND DISCUSSION

5.1 *Locally generated ozone transported within St. Louis*

Ozone concentrations were categorized with respect to local-scale trajectories either intersecting or missing HEAs during the days when the NAAQS for ozone was exceeded at any of the five outer sites(Fig. 3). The same information in Fig. 3 is summarized in a contingency format in Table 1. In this table it is clear that the highest ozone concentrations were more frequently associated with air that had just traversed a HEA.

Data from August 19, 1975 substantiates the consequence of the local transport of ozone generated within the HEAs of St. Louis. The highest ozone concentration measured at any of the RAMS stations was observed at station 121 on this day. Between 1500

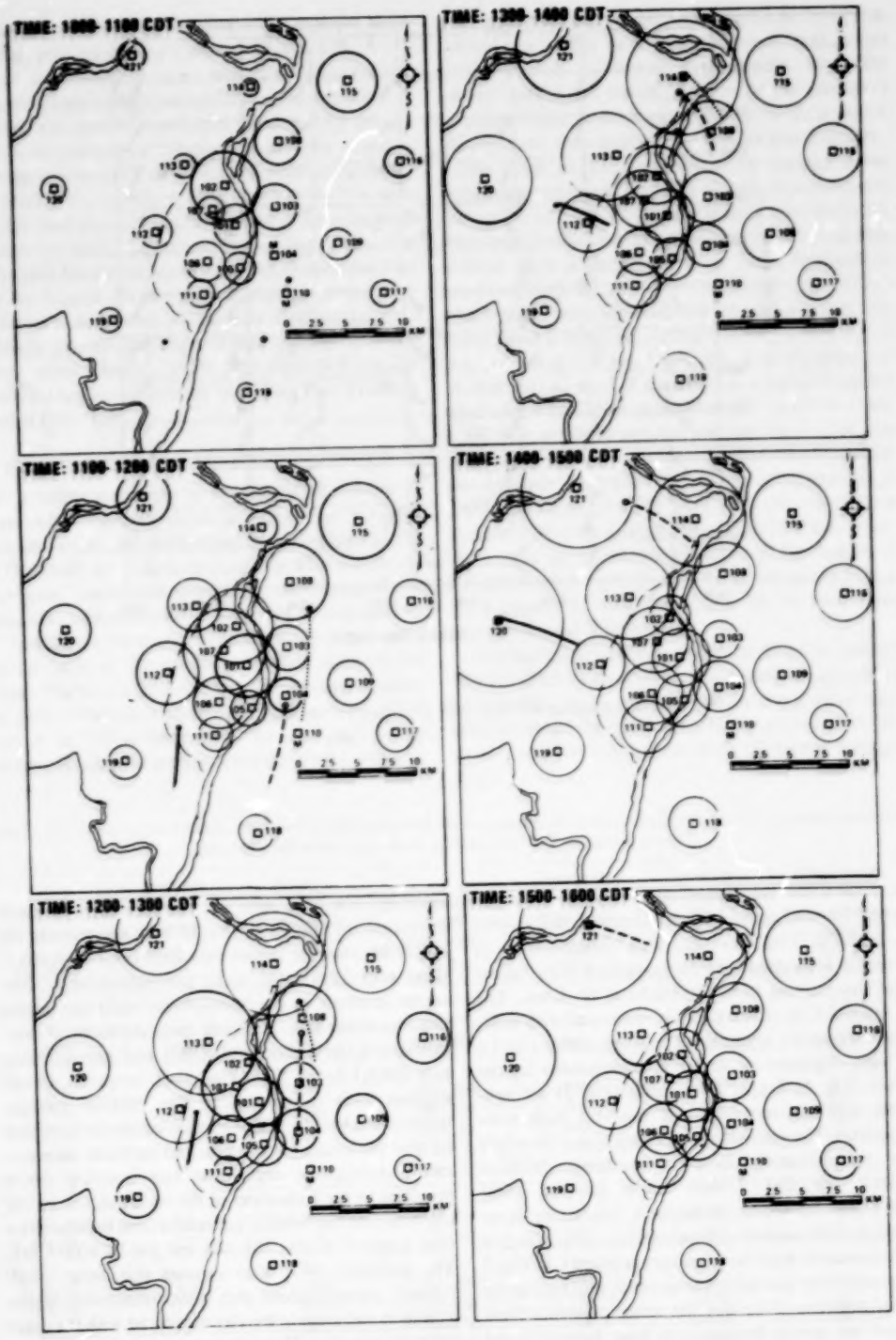
TIME: 1000- 1100 CDT
TIME: 1200- 1400 CDT
TIME: 1100- 1200 CDT
TIME: 1400- 1500 CDT
TIME: 1200- 1300 CDT
TIME: 1500- 1600 CDT
0 2.5 5 7.5 10 KM
CONVERSION CIRCLE
= [O3] OF 80 ppb

Fig. 4. Time sequence of ozone concentrations in the RAMS for August 19, 1975. Trajectory segments are shown which represent the one-hour movement of the air which arrived at stations 114, 120 and 121 during the hour of maximum ozone concentration at these stations.

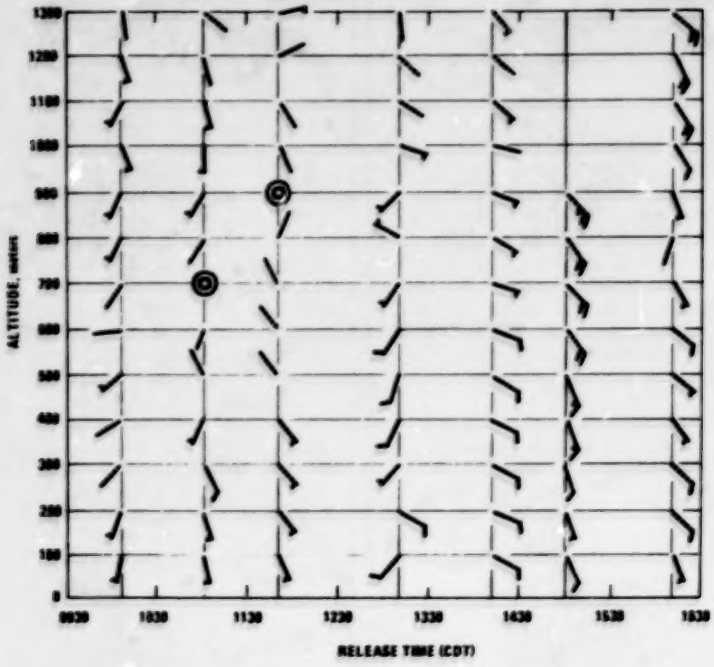

Fig. 5. Time sequence of winds aloft at station 141 on August 19, 1975.

and 1600 CDT the ozone concentration averaged 233 ppb ($\simeq 430\,\mu g\,m^{-3}$) approximately three times the NAAQS. This unusually high concentration of ozone was associated with the movement of an "ozone cloud" originating from the HEAs of St. Louis. The movement of the ozone cloud is illustrated with near-surface trajectory segments at selected stations and a 6 h time sequence of ozone concentrations in the RAMS (Fig. 4). Upper level winds (Fig. 5) are consistent with the movement of the very high concentrations. Throughout the morning hours the winds blew predominantly from a southerly direction at least the first 1300m above ground. Near 1300 CDT the winds throughout this entire layer shifted to more easterly and increased in speed which is quite consistent with the trajectory segments. In Fig. 4 it is noteworthy that for stations south and east of the central portions of the city the ozone concentrations did not vary greatly from hour to hour, but north and west of the city center the ozone concentrations varied markedly from hour to hour. During the hours (1000–1200 CDT) when ozone rapidly increased in concentration at sites 101, 102, 107, 108, 112, 113 and 115 the concentration at all other sites remained relatively low. This rules out the possibility of ozone advection into this area, but it does not exclude the possibility that precursors may have been advected to these sites from other areas and subsequently converted to ozone by the intense solar radiation during this cloud-free day. Ambient measurements of non-methane hydrocarbons (NMHC) and nitrogen dioxide (NO_2) in the RAMS network however, do not support such a contention. The NMHC concentrations at sites 116–125 remained below the detection level of the total hydrocarbon and methane measurements during the night-time and morning hours previous to the formation of the ozone cloud. In the city however, the NMHC concentrations ballooned up over 1 ppm C at sites 101, 103 and 104 by 1000 CDT. The pollutant NO_2 also showed this same trend, namely, concentrations that were consistently higher at sites in and close to the city compared with the outer surrounding sites. The evidence strongly suggests that the ozone cloud was locally generated and its origin was near sites 101, 103 and 104. The movement of the ozone cloud out from the inner areas of St. Louis was the obvious cause for such high readings north and west of St. Louis. Unfortunately, the cloud could not

be traced beyond site 121 because all the ozone data for the day were missing at stations 122 and 125.

It is interesting to note that the highest ozone concentrations on this particular day did not occur in the city, but were downwind of the city. This phenomenon is partially attributed to the fast reaction between ozone and locally emitted nitric oxide in the city ($NO + O_3 \rightarrow NO_2 + O_2$). In addition, it is conceivable that on a substantial number of days the wind advects the ozone precursors out of the urban area before they have a chance to react and form ozone. Not only on August 19, but on the average, ozone concentrations downwind of the city tend to be substantially greater than those within the city. For this reason rural stations (121–125) have statistically significant higher ozone concentrations than do urban stations (101–107).

5.2 *Ozone transported into St. Louis*

It is rather surprising to see the large number of times ozone concentrations exceeded the NAAQS when the local-scale trajectories missed the HEAs (Fig. 3). Therefore the degree of accuracy with which the local-scale trajectories represent the transport of ozone may be legitimately questioned. It is subject to two conditions. First and foremost is the assumption that the bulk of the ozone is confined close to the surface. Numerous investigators (Polgar and Londergan, 1976; Lovelace *et al.*, 1974; Ripperton *et al.*, 1976; Lusis *et al.*, 1976) have observed that high ozone concentrations near ground level drop off rapidly to low concentrations above the subsidence inversion. Second, on some days the computed trajectories probably had significant discrepancies from the airflow within the lower portions of the planetary boundary layer because of wind shear in these layers. PIBAL data were analyzed to resolve this difficulty.

The weighted range and the vector average of the wind were calculated from the PIBAL measurements during the 12 h previous to the start time of the local-scale trajectories. The calculations included wind data at every 50 m beginning at 100 m. These results were used to exclude from further consideration those days which may have been affected by air that had just passed over a HEA. An example of the application of this procedure follows. Using Fig. 1 as a guide, it is evident that at site 121 the directions from which the wind misses the HEAs range from 235° to 40°. On July 4, 1975 the weighted range of the wind during the 12 h previous to the time the NAAQS had been exceeded was between 282° and 355° (Table 2). Since this is within the range from 235° to 40° the wind would not transport air from a HEA to station 121. Table 2 lists those days on which one or more of the five rural RAMS sites were not affected by the emissions from a HEA based on both the results of the local-scale trajectories and PIBAL data.

The day previous to each of those listed in Table 2 was checked to see if the wind direction had shifted so significantly that rural stations could have been affected by air that had recently flowed out of the St. Louis area. In all but three cases the vector average

Table 2. Wind data at the PIBAL sites for 12 h previous to the time of maximum ozone concentration on days which were not under the influence of air flowing from the "high emission areas"

DATES	Average wind speed ($m\,s^{-1}$) ($\bar{V}$)	Resultant wind direction (degrees) ($\bar{V}$)	Weighted range (degrees) (WR)	Number of 50 m levels dimensionless (N)	Average height of PIBALS (m) (H)	Maximum ozone concentrations (ppb) [O_3]
SITE 121						
7/4/75	8.7	318	282–355	222	1550	92
9/2/75	10.0	331	302–360	665	1000	88
SITE 122						
7/4/75	8.8	320	282–358	198	1550	85
5/21/76	6.5	286	242–330	701	1450	100
6/1/76	3.7	6	290– 81	440	850	98
6/2/76	9.0	38	6– 71	290	800	82
6/3/76	8.9	53	19– 87	643	1400	86
SITE 123						
5/27/76	8.3	117	49–184	605	1400	81
6/2/76	6.9	36	5– 67	291	700	81
SITE 124						
8/28/75	4.9	180	109–251	600	1450	81
5/5/76	21.1	210	204–256	750	1550	88
5/26/76	6.9	117	81–154	769	1400	88
6/10/76	10.5	223	205–240	508	1450	97
6/11/76	7.3	243	187–299	788	1400	91
6/17/76	6.9	194	134–252	689	1450	81
SITE 125						
8/10/75	7.7	214	169–259	1095	1400	86
6/9/76	4.8	237	187–286	613	1400	88

wind at each 50 m level, beginning at 50 m, would not have crossed any portion of a HEA. For the other three cases, June 1, 1976; May 26, 1976; and September 2, 1975, the 24 h vector average wind on the previous day barely brushed the edge of a HEA, but only for two of the 50 m incremented levels. Clearly, a 24 h return flow was not the case during any of these days.

The lack of local anthropogenic emissions of NO_x and THC in the immediate vicinity of the rural RAMS sites preclude the possibility of local sources near these stations as the cause for such high ozone concentrations. At stations 122–125 the local emissions of NO_x and THC for a radius of 15 km are approx one-hundredth of the emissions within a 15 km radius of any of the urban stations. Of course, this would not be valid for station 121 because a circle would enclose sources of high NO_x and THC emissions located within the HEAs. Nonetheless, the local emissions of THC and NO_x north and west of station 121 are quite small when compared to the emissions around any of the urban sites. For example, the NO_x and THC emissions within a semi-circle of radius 15 km with its diameter centered on station 121 are approx 5% of the emissions within a circle of 7.5 km radius surrounding any of the urban sites.

Large-scale trajectories are shown for the days of interest (listed in Table 2) in Fig. 6. The starting times of these backward trajectories were either 1300 or 1900 CDT, depending on the time of the maximum ozone concentration at the outer sites. The accuracy associated with Heffter's large-scale trajectories is currently being investigated within the meteorological community, but if the results of a recent study by Hoecker (1977) can be generalized, an average error of 15–20 km can be expected with each 100 km segment of these trajectories.

It is important to note that the trajectory paths are not limited to any specific quadrants. Observational data (Angus and Martinez, 1974; Ripperton *et al.*, 1976) have led investigators to conclude that a comparatively high value of the diurnal maximum ozone concentration is often associated with the back side of high pressure systems. As Ripperton *et al.* (1976) explain, this would allow time for the accumulation of ozone, the formation of ozone from slow reacting precursors, and the buildup of a large or critical concentration of ozone precursors. Table 3 reveals that indeed, for the vast majority of the days listed in Table 2 the air entering St. Louis had been dominated by a high pressure system for about 3 days prior to its arrival into the area. This was determined by noting the pressure pattern along the trajectories beginning in St. Louis. The air was not considered as being under the influence of a high pressure system when the curvature of the surface isobars was no longer anticyclonic. The National Oceanic and Atmospheric Administration's *Daily Weather Map Series* was utilized for this purpose.

It is not surprising to find anticyclones dominating the airflow entering St. Louis for nearly all of the days listed in Table 1. The dynamics of high pressure systems are such that the movement of air within them is usually relatively slow, but nevertheless is sufficiently fast enough to transport ozone over several hundred kilometers within 24 h. Also anticyclones, especially those with a warm core, frequently move quite slowly or even become stationary (Korshover, 1975). When these conditions occur over the eastern half of the United States where numerous high intensity urban-industrial centers exist, conditions are conducive not only for ozone generation, but also for large-scale ozone transport.

Some comments are necessary regarding 2 days when the NAAQS for ozone was exceeded and a high pressure system did not dominate the airflow previous to its arrival into St. Louis. On these 2 days a weak trough of low pressure and a stationary front were situated near the St. Louis area. During these days trajectories indicate that the air flowing into St. Louis passed within 75 km of the very large Chicago–Gary urban-industrial complex just 12–24 h earlier. On these two days it is likely that this area was responsible for the high ozone concentrations in St. Louis. Others (Wilson *et al.*, 1976) have observed that urban plumes of ozone travel hundreds of kilometers from areas only half as populated as the Chicago–Gary area.

Stratospheric sources of the high ozone concentrations have not been considered up to this point. While it is conceivable that on occasions stratospheric intrusions of ozone can cause surface ozone concentrations above the NAAQS, Reiter (1976) and Singh *et al.* (1975) conclude that ozone concentrations in excess of the NAAQS can occur only with a combination of two conditions. First, there must be cyclogenesis in the troposphere with a pronounced tropopause folding event and secondly, there must also be a strong planetary wave trough present in the middle and lower stratosphere. Due to the dominance of anticyclones associated with the trajectories, not

Table 3. The number of days that air had been dominated by surface high pressure previous to its arrival into St. Louis on the date listed

Dates of high $[O_3]$	Number of days
7/4/75	≥ 3
8/10/75	≥ 3
8/28/75	≥ 3
9/2/75	≥ 3
5/5/76	2
5/21/76	≥ 3
5/26/76	≥ 3
5/27/76	≥ 3
6/1/76	0
6/2/76	0
6/3/76	≥ 3
6/9/76	≥ 3
6/10/76	≥ 3
6/11/76	≥ 3
6/17/76	2

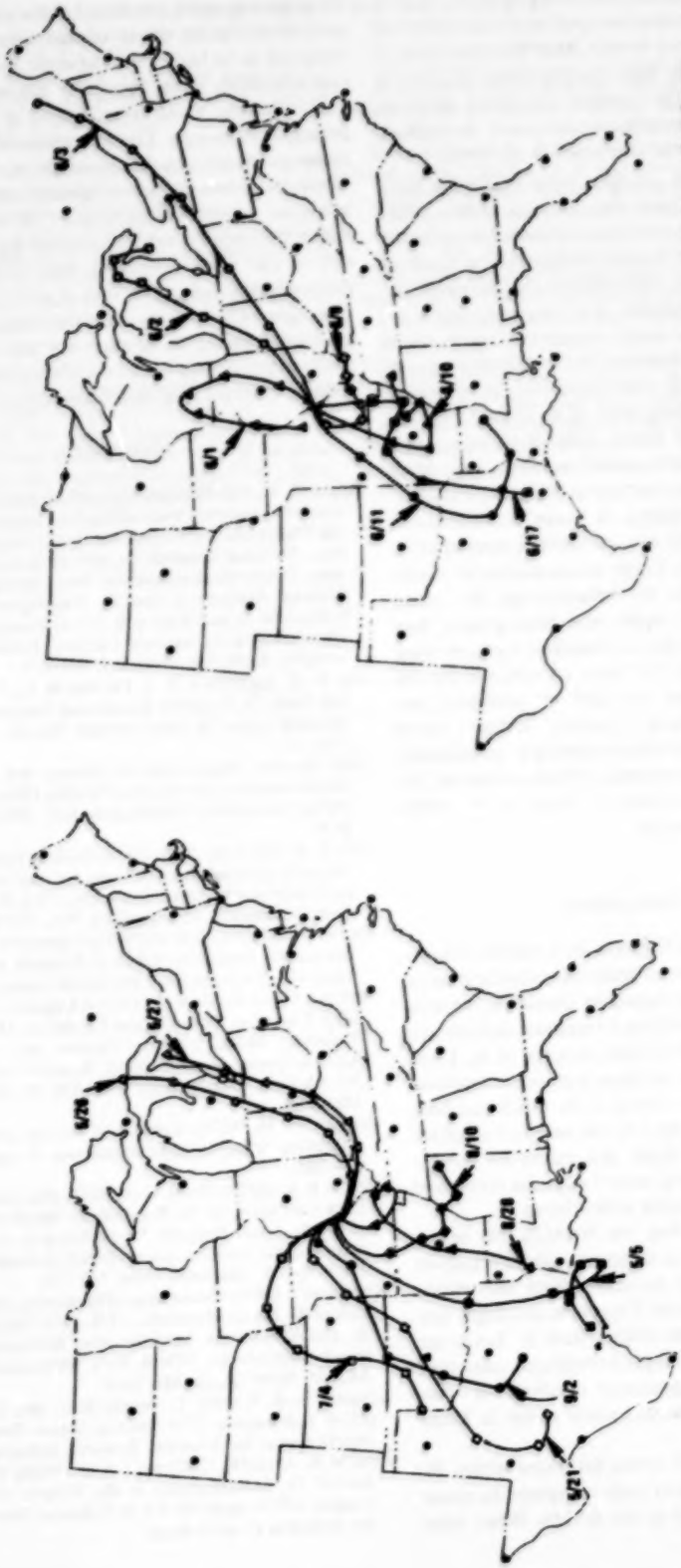

Fig. 6. 72 h backward in time trajectories which begin in St. Louis for the dates indicated. Each 12 h segment can be noted by the spacing of the various symbols along each trajectory.

even the first criterion was fulfilled. Even if it is assumed that stratospheric injection of ozone into the troposphere occurred remote from the trajectories it would be difficult for high concentrations of ozone to penetrate beneath the synoptic subsidence inversion which occurs frequently in stationary as well as travelling anticyclones (Pettersen *et al.*, 1946).

Emissions of hydrocarbons from vegetation have been suggested by some (Stasiuk and Coffey, 1974; Saunder *et al.*, 1974) as a means of contributing to the formation of ozone. Recent measurements however (Gay and Arnts, 1976, 1978), suggest that the emissions of natural hydrocarbons (e.g. terpenes) have no significant effect on ozone concentrations in rural areas. In addition, because of the contrasting types of vegetation underlying some trajectories as compared to others (*The National Atlas of the United States of America*, 1970) it is rather difficult to argue that naturally emitted hydrocarbons were the cause of all the high ozone concentrations at the rural stations.

In reality the frequency of ozone concentrations exceeding the NAAQS at rural stations upwind of St. Louis, where the St. Louis urban-industrial center would have little or no influence on the ozone concentrations, could easily have been greater than cited. The reasons for this are threefold. First, the days which were considered to have no urban-industrial influence were judged as such by stringent meteorological procedures; second, surface ozone measurements were sometimes missing or invalidated; and third, on most weekends, which accounted for nearly 20% of the number of days in the study, PIBALs were not released.

6. CONCLUSIONS

Using data from the RAPS in St. Louis both large-scale and local transport of ozone were shown to occur frequently. Ozone concentrations above the NAAQS at rural locations occurred on 15 separate days when it was clear the urban-industrial complex of St. Louis had very little influence on these high concentrations. On these days the air arriving in St. Louis had been confined for at least 3 days to the eastern half of the United States where there are numerous urban-industrial centers. During these 3 days the movement of air was contained within anticyclones.

The results suggest that the NAAQS will be exceeded in St. Louis due to large-scale ozone transport regardless of precursor emissions from the urban-industrial areas of St. Louis. The results also imply that the precursors and ozone emitted from St. Louis can be, under certain meteorological conditions, combined with significantly high precursor emissions in other areas and contaminate areas outside of the St. Louis region.

The local transport of ozone generated within St. Louis must be considered in order to explain the ozone concentrations measured in the RAMS. When rural areas were situated downwind of the urban-industrial area of St. Louis, ozone concentrations were often observed to be higher than at other times. In fact the concentrations were frequently higher than urban concentrations which were located in areas of high precursor emissions. This was exemplified by noting on one particular day the movement of a surface-based cloud of ozone which originated in the urban-industrial complex of St. Louis. The concentrations within the ozone cloud increased as the cloud moved out of the urban-industrial area and resulted in extremely high concentrations at outer sites.

In light of the results from this study it would be most appropriate to develop control strategies for ozone which consider both the consequences of large-scale ($\geq$ 200 km) and local transport of ozone.

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